ANTIFERROMAGNETIC TO PARAMAGNETIC TRANSITION IN Fe^{2+} : MnF_2 IN EXTERNAL MAGNETIC FIELDS

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ABSTRACT

Mössbauer effect measurements across the antiferromagnetic tc paramagnetic phase boundary in Fe^{2+}: MnF_2 in external magnetic fields are reported. From the data, J(Mn-Fe)= -1.7 cm^{-1}. The phase boundary is found to vary as H_S^2, as in pure MnF_2.

MnF_2 crystallizes in a rutile structure with a tetragonal lattice. Below the Néel temperature T_N=67.4K, the magnetic properties of MnF_2 are well understood in terms of an ideal, two sublattice antiferromagnet with the spins aligned along the tetragonal c-axis. The phase diagram of MnF_2 in the H-T plane has been studied by Shapira and Foner[1] and is shown in Fig. 1. At low temperature, an external magnetic field applied along the c-axis causes a first-order realignment of the sublattice magnetization from along the c-axis to the basal plane when the magnitude of the external field reaches the critical value H_{sf}. At higher temperature, specifically above 65 K, the external field causes a second order transition to the paramagnetic phase, i.e., effectively lowers the Néel temperature.

Fe^{2+} may be isomorphously incorporated into the MnF_2 lattice. The effect of the addition of iron is to increase the Néel point[2] and to increase the value of H_{sf}[3]. In a previous work, we observed the spin flop in Fe^{2+}: MnF_2 using Mössbauer spectroscopy in ^{57}Fe[4]. In this paper we report measurements of the antiferromagnetic to paramagnetic phase transition in external magnetic fields for single crystals of MnF_2 doped with 1% ^{57}Fe.

A large single crystal of ~ 1% $^{57}Fe^{2+}$ doped MnF_2 was grown from the melt by Optovac, Inc. The single crystal was oriented and a 6 mil slice was cut perpendicular to the c-axis and mounted between beryllium disks. In addition, some of the crystal was crushed and the powder was cast in lucite. Measurements were made in a conventional constant acceleration spectrometer operating in the normalized mode. The sample was placed in a cryostat which was inserted in a liquid nitrogen dewar which was in turn inserted into a superconducting solenoid operating in the persistent mode up to 85 kOe. The temperature was

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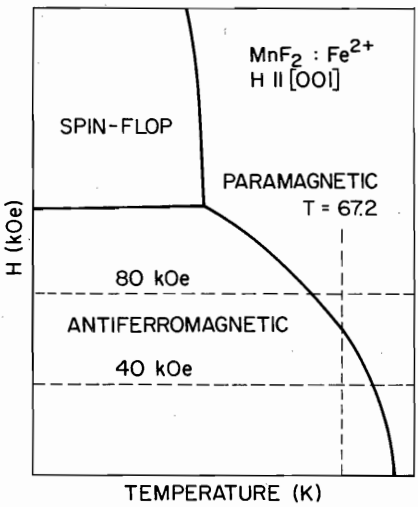

Fig. 1.

Phase diagram of pure MnF_2 from Ref. 1. Inclusion of Fe^{2+} raises H_{sf} and increase T_N. The measurements reported here are across the AF-P phase boundary.

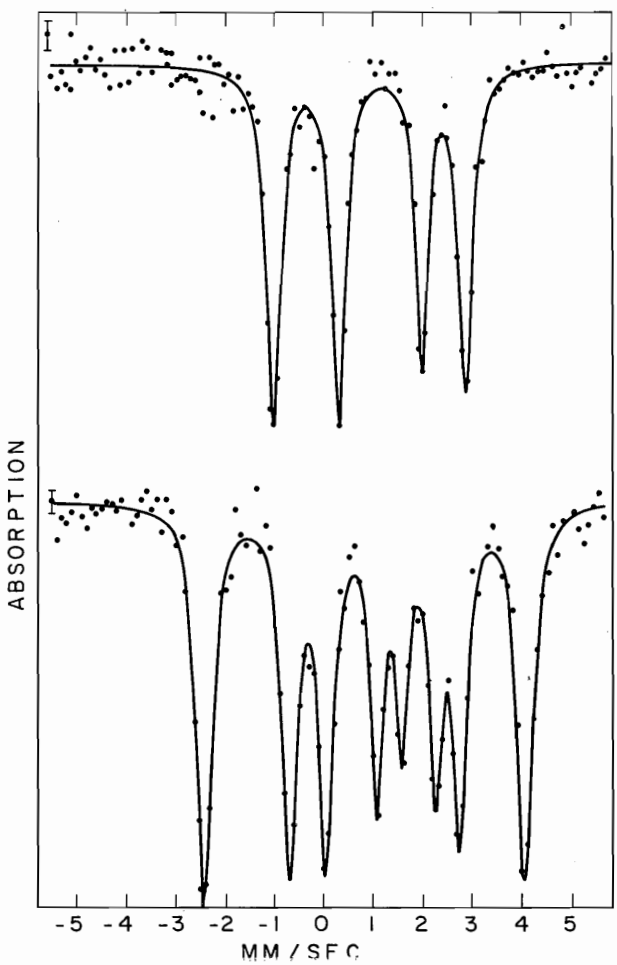

Fig. 2.

Mössbauer spectra of single crystal Fe^{2+}: MnF_2 with H_o= 80 kOe along the c-axis above and below T_N.

controlled by pumping the liquid nitrogen bath below the desired temperature and then heating the sample electrically. The temperature was measured using a wire wound Pt resistor and the values were corrected for the effect of the external field using the results of Neuringer et al.[5]

Because the field at the nucleus $\vec{H}_n = \vec{H}hf \pm \vec{H}_o$, the spectra of the spin up and spin down sublattices in the antiferromagnetic phase are observed independently and the complete spectrum consists typically of eight lines (Fig. 2). In the paramagnetic phase all the spins are equivalent and the spectrum consists of just four lines (the $\Delta m = 0$ lines are absent because the γ-ray propagation direction is parallel to H_o). Thus we observe the temperature dependence of the spin up and spin down sublattices independently and the transition to the paramagnetic phase in clearly delineated.

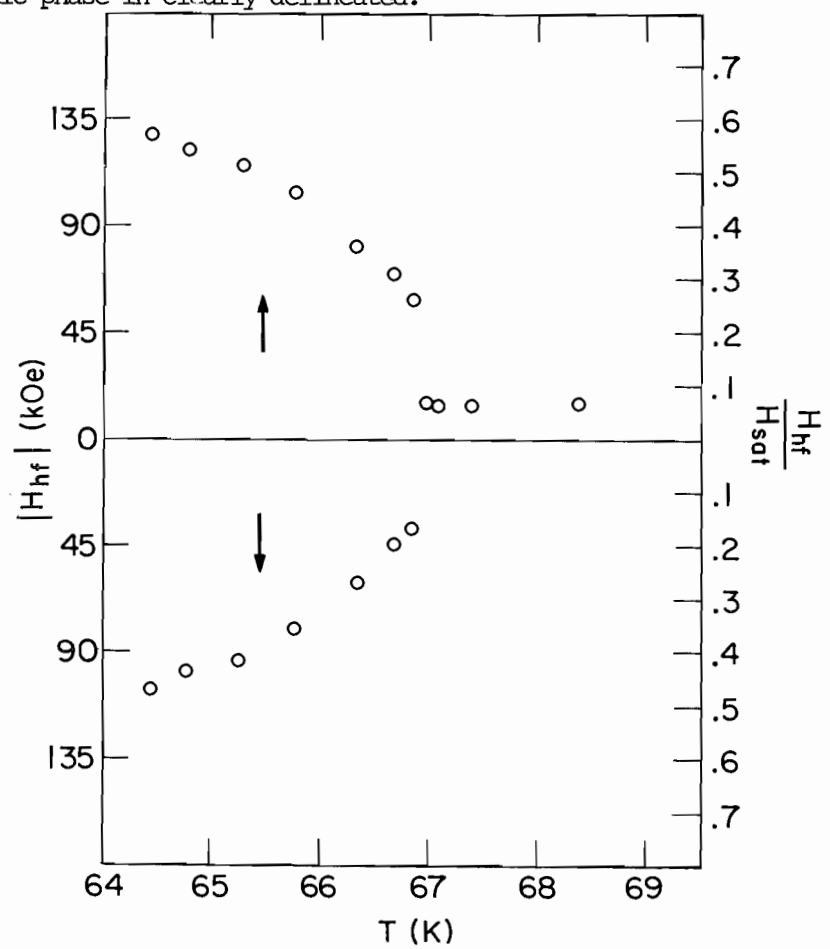

Fig. 3. H_{hf} and Hhf/H_{sat} plotted as a function of T for H_o=80 kOe.

The data were analyzed using a computer program of Singh and Hoy[7]. From the quadrupole splitting above T_N and powder spectra at several points below T_N we found that the quadrupole coupling parameter $Q = 2.95$ mm/sec. and the asymmetry parameter $\eta = 0.4$ did not change appreciably in the transition region.

In Fig. 3. we plot the hyperfine field H_{hf} in the Fe^{2+} in the spin down and spin up sublattices as a function of temperature for an applied field $H_o = 80$ kOe. The data have been analyzed using a molecular field approximation model wher J(Mn) and J(Fe), the Mn-Mn and Fe-Fe exchange constants respectively were chosen to give the best fit to the Néel point of pure MnF_2 and pure FeF_2. The Fe^{2+} single ion anisotropy constant D was taken from the measurements of Lowe et al;[3] $D = 8.6$ cm^{-1}. Using these values the best fit to the Néel point is obtained with $J = -1.5$ cm^{-1}, however the best fit to the magnetigation above and below T_N is obtained with $J = -1.7$ cm^{-1}.

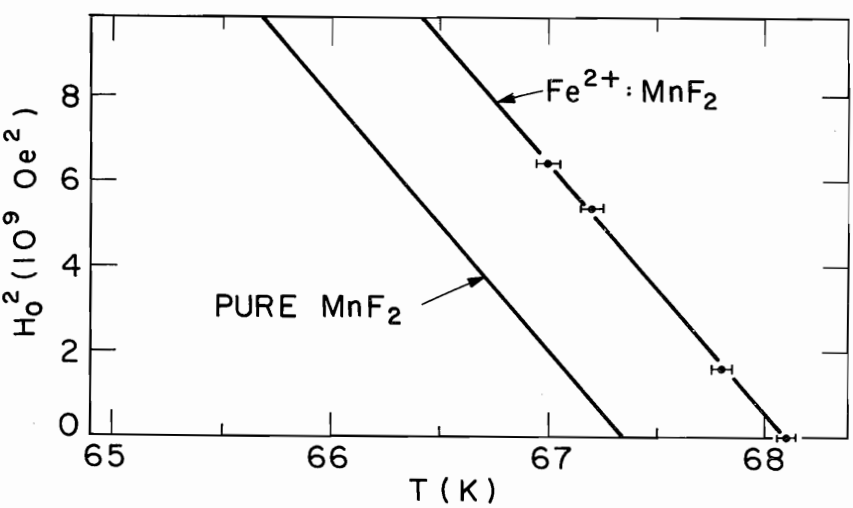

Fig. 4. H^2 plotted as a function of T for MnF_2 and $Fe^{2+}:MnF_2$

Shapira and Foner[1] and Heller[6] showed that the phase boundary in pure MnF_2 is well represented by an equation of the form $T - T_N = AH^2$, where A is a constant. In Fig. 4 we plot T_N determined from our data as a function of H_o^2, including one value obtained by holding T constant and changing H_o. The boundary is seen to vary as H_o^2 as in MnF_2 and with the same slope. This result is predicted by the M.F.A.

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